

### REMARKS

Claims 1-21 are pending. Claims 22-28 are canceled.

Claims 1-21 stand rejected under 35 USC § 112 as being indefinite because the phrases "substantially free of," and "incompletely oxidized reaction products comprising" are recited. For the reasons which follow, Applicants submit that these phrases are definite.

The phrase "substantially free of" is not uncommon language. The phrase "substantially free of" is less than an amount of zirconium which increases solvent burn, and in this case, which increases solvent burn by more than 10 % relative to the same catalyst composition devoid of zirconium. Support for this definition may be found on page 8, lines 15-29.

"Reaction products comprising 4-carboxybenzaldehyde compounds" are definite because "comprising" modifies "reaction *products*" rather than a reaction *product* in the singular. There can be a variety of incompletely oxidized reaction products present, among which is at least 4-carboxybenzaldehyde compounds. Comprising is indeed an open-ended term, and in this case the word comprising calls for at least one of the many products to be 4-carboxybenzaldehyde compound. It is submitted that this phrase is definite and clear. For these reasons, Applicants respectfully request withdrawal of the rejection of claims 1-21.

Claims 1-16 and 21 are rejected under 35 U.S.C. 103(a) as being unpatentable over Park et al., U.S. 6,476,257 ("Park et al.") in view of Shirgeyasu et al., US 4,160,108 ("Shirgeyasu et al."). For the following reasons, Applicant respectfully traverses the rejection.

Claim 1 calls for, among other things, a process for the oxidation of p-xylene to terephthalic acid comprising a nickel, manganese, and bromine catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and the amount of nickel atoms is at least 500 ppm. Claim 1 is patentable over Park et al. in view of Shigeyasu et al. because neither reference alone or in combination discloses or suggests a process for the oxidation of p-xylene to terephthalic acid comprising a nickel, manganese, and bromine catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and the amount of nickel atoms is at least 500 ppm.

Park et al. is directed to adding nickel and carbon dioxide in appropriate amounts to increase the activity of cobalt/manganese/bromine complex catalyst in the oxidation of alkylaromatics to form aromatic carboxylic acids. (Col.1, lines 11-22). Park et al. do not disclose a process for the oxidation of p-xylene to terephthalic acid comprising a nickel, manganese, and bromine catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and the amount of nickel atoms is at least 500 ppm. In fact, the examples disclosed in Park et al. use 899 ppm cobalt, 1170 ppm manganese and 2990 ppm for bromine. (Col. 4, lines 9-13, and all following examples). The stoichiometric molar ratio of bromine atoms to manganese atoms is 1.76, not 1.5 or less. Also the amount of nickel used as a catalyst in the examples is 163 ppm of nickel based on the total weight of reactants which corresponds to 28 ppm based upon total weight of p-xylene and acetic acid feed. (Col. 4 lines 7-18). The 28 ppm of Nickel is far below Applicant's Claim 1 wherein the catalyst comprises nickel atoms of at least 500 ppm based on the weight of all liquid and solid feeds.

Shigeyasu et al. is directed to a process for producing high purity terephthalic acid suitable for use as starting materials to obtain a high quality polyester by direct polymerization with glycols. (Col. 1 lines 8-12). Shigeyasu et al. disclose use of a catalyst system comprising distinct, temperature-dependent ranges of cobalt compound composition and bromine compound composition as well as two requirements for the

amount of manganese used (1) the weight ratio of the manganese component to the cobalt component in the catalyst is 0.25:1 to 1:1 and (2) the amount of the manganese component is also 0.05% by weight or less based on the weight of the lower aliphatic carboxylic acid solvent. (Col. 3 line 55 – Col. 4 line 67). Shigeyasu et al. do not disclose a process for the oxidation of p-xylene to terephthalic acid comprising a nickel, manganese, and bromine catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and the amount of nickel atoms is at least 500 ppm.

The catalyst system disclosed by Shigeyasu et al. does not have a stoichiometric molar ratio of bromine atoms to manganese atoms of 1.5 or less nor an amount of nickel atoms of at least 500 ppm. Shigeyasu et al. disclose a range of Bromine wt % in the solvent of 0.112 to 0.7 and a maximum manganese wt % in the solvent of 0.05. (Col. 4 lines 15-21 and lines 56-63). These values give a corresponding minimum stoichiometric molar ratio of bromine atoms to manganese atoms of 1.54 to 9.62 (note that considering a minimum amount of manganese, the stoichiometric molar ratio can be as high as 34). Shigeyasu et al. disclose that a promoter of bromide with any of eighteen different metals, including nickel, may be added, but such is not essential. (Col. 6, lines 50-59). Shigeyasu et al. further disclose that when added, the amount of such metal compound, calculated as elemental metal, added is generally 0.1 to 10 wt % based on the weight of cobalt. (Col. 6, lines 65-68). Again, using the formula of Shigeyasu et al. for the amount of cobalt and maximizing the amount of nickel at 10 wt% based upon Cobalt, Nickel can be 50 ppm to 175 ppm based upon the weight of the solvent (Col. 3, lines 55-60). Applicant's catalyst comprises greater than 500 ppm Nickel based upon total liquid and solid feeds.

In sum, neither Park et al. nor Shigeyasu et al., individually or in combination, disclose or suggest at least two elements of Applicant's claimed invention: a stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and at least 500 ppm Nickel. Applicant's respectfully note that a prima facie case of

obviousness which requires that the prior art references when combined must teach or suggest all claim limitations has not been met since neither Park et al. nor Shigeyasu et al. nor the combination of the references teach or suggest a process for the oxidation of p-xylene to terephthalic acid comprising a nickel, manganese, and bromine catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less and the amount of nickel atoms is at least 500 ppm.

Claims 1-21 are rejected under 35 U.S.C. 103(a) as being unpatentable over Partenheimer et al., US 4,786,753 ("Partenheimer et al.") in view of Shigeyasu et al., US 4,160,108 ("Shigeyasu et al."). For the following reasons, Applicant respectfully traverses the rejection.

Claim 1 calls for, among other things, a process for the oxidation of p-xylene to terephthalic acid comprising a catalyst composition substantially free of zirconium comprising nickel, manganese, and bromine wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less. Claim 1 is patentable over Partenheimer et al. in view of Shigeyasu et al. because neither reference discloses or suggests a process for the oxidation of p-xylene to terephthalic acid comprising a catalyst composition substantially free of zirconium comprising nickel, manganese, and bromine wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less.

Partenheimer et al. is alleged to expressly teach "the method of producing terephthalic acid by oxidizing p-xylene in the presence of a catalyst composition comprising Ni, Mn, and bromine at a pressure of 150 psig and at a temperature of 160° C." This is incorrect for the reasons which follow.

Partenheimer et al. suggest, and even require, the presence of zirconium in the catalyst system in order to improve the oxidation rate of p-xylene, m-xylene and pseudocumene. Partenheimer et al. would not have suggested the claimed invention

because the claimed invention calls for a catalyst system that is substantially free of zirconium atoms. This feature is not suggested by Partenheimer et al., and in fact, Partenheimer et al. teach against a catalyst system that is substantially free of zirconium atoms.

Furthermore, Partenheimer et al. do not suggest or disclose a catalyst wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less. Table II of Partenheimer et al., from which the alleged express teaching of "the method of producing terephthalate acid by oxidizing p-xylene in the presence of a catalyst composition comprising Ni, Mn, and bromine at a pressure of 150 psig and at a temperature of 160° C." is taken has several errors and inconsistencies. Specifically, footnote "a" underneath Table II, describes the reaction conditions for Examples 15-18, refers to Example 17 and 18 as oxidizing 15.0 ml p-xylene, and Examples 19 and 20 as oxidizing 15 ml m-xylene. This cannot be correct because Examples 19 and 20 are not in Table II and actually fall under Table III, which describes the oxidation of pseudocumene. It is possible that what was actually intended was to describe Examples 15 and 16 as an oxidation in 15 ml of p-xylene, and Examples 17 and 18 as an oxidation in 15 ml of m-xylene.

Another error noted in Table II lies with respect to the amount of bromine identified in Example 17 as 2.01 moles. It is believed that the number of "2.01" is a typographical error because at Column 4, lines 24-28, the Patentee notes that:

It is therefore important when determining if a synergistic interaction has occurred, to be sure that *equal catalyst* concentrations are being compared. In the following examples this proviso *has been observed*. [Emphasis added].

Further, it was also noted that:

"in Table I the experimental data as set forth where pseudocumene was the oxidant while in Table II p-xylene and m-xylene were the oxidants. The synergistic effect of zirconium on Ni/Mn/Br catalyst was observed with all of these feedstocks." Column 4, lines 46-50.

Thus, according to Partenheimer et al., the catalyst concentration across Examples 15-18 should have been kept the same in order to evaluate whether zirconium did or did not have a synergistic effect on the oxidation rate. Specifically, footnote "a" underneath Table II states that

"Reactions [examples 15-18] were run in a glass reactor containing 2.01 mmole cobalt (II) acetatetetrahydrate, 2.01 mmole manganese (II) acetatetetrahydrate, 4.00 mmole sodium bromide, and 100.0 ml. acetic acid."

For this reason, it is believed that those of ordinary skill, reading Example 17, would have believed that the designation of 2.01 moles for the bromide was an error, and should have been stated as 4.01 as in Examples 15, 16, and 18. The corresponding stoichiometric atomic ratio of bromine to manganese in Examples 15-18 is 2.0, higher than Applicant's claim element of 1.5 or less.

Partenheimer et al. disclose a stoichiometric atomic ratio of total metals, nickel plus magnesium plus zirconium, to bromine in the range of about 0.5 to about 1.5. (Col. 2 lines 6-9). The corresponding stoichiometric range of bromine to all metals is 0.67 to about 2.0. There is no motivation or suggestion within Partenheimer et al. to consider the ratio between bromine and manganese. If one does combine Partenheimer et al.'s disclosure of stoichiometric ratio of nickel-zirconium-manganese in the range of 33:1:12 to about 80:1:43, the corresponding calculated stoichiometric ratio of bromine to manganese is 1.9 to 7.7, well above Applicant's claim element of a stoichiometric ratio of bromine to manganese of 1.5 or less.

Thus, the deficiencies in Partenheimer et al. are not only with respect to the content of 4-CBA, or the claimed ratio of solvent burn, but also in that Partenheimer et al. do not suggest a process for the oxidation of p-xylene to terephthalic acid comprising a catalyst composition substantially free of zirconium comprising nickel, manganese, and bromine wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less. Moreover, for the reasons noted above with respect to

the deficiencies of Table II, it is doubtful that those of ordinary skill in the art would have been informed by Example 17 that the stated amount of bromine is correct.

Finally, it must be borne in mind that the only teaching relied upon to support an obviousness rejection is a *comparative* example, and that the only teaching within Partenheimer et al. suggesting a modification of a comparative example to improve the oxidation rate is to employ zirconium atoms. The fact that Example 17 is a comparative example is powerful evidence pointing away from and teaching against the use of a Ni/Mn/Br catalyst system substantially free of zirconium atoms, and does not provide any teaching whatsoever on how to modify that composition catalyst system in a manner other than by addition of zirconium atoms.

It is acknowledged in the Office Action that the prior art is silent about the solvent burn ratio, but it is alleged that analyzers can be used to measure the degree of burn according to Partenheimer et al. However, it is submitted that the mere existence of capabilities to measure the degree of acid burn does not suggest what the ratio of acid burn should be, or the operating conditions that should be used to lower that ratio, or what the operating conditions in combination with a particular catalyst composition should be to obtain optimally lower solvent burn, especially in combination with lowered amount of 4-CBA as set forth in Applicant's Claims 20 and 21.

Shigeyasu et al. is directed to a process for producing high purity terephthalic acid suitable for use as starting materials to obtain a high quality polyester by direct polymerization with glycols. (Col. 1 lines 8-12). Shigeyasu et al. disclose use of a catalyst system comprising distinct, temperature-dependent ranges of cobalt compound composition and bromine compound composition as well as two requirements for the amount of manganese used (1) the weight ratio of the manganese component to the cobalt component in the catalyst is 0.25:1 to 1:1 and (2) the amount of the manganese component is also 0.05% by weight or less based on the weight of the lower aliphatic carboxylic acid solvent. (Col. 3 line 55 – Col. 4 line 67). Shigeyasu et al. do not

disclose a process for the oxidation of p-xylene to terephthalic acid comprising a catalyst substantially free of zirconium comprising nickel, manganese, and bromine wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less.

The catalyst system disclosed by Shigeyasu et al. does not have a stoichiometric molar ratio of bromine atoms to manganese atoms of 1.5 or less. Shigeyasu et al. disclose a range of Bromine wt % in the solvent of 0.112 to 0.7 and a maximum manganese wt % in the solvent of 0.05. (Col. 4 lines 15-21 and lines 56-63). These values give a corresponding minimum stoichiometric molar ratio of bromine atoms to manganese atoms of 1.54 to 9.62 (note that considering a minimum amount of manganese, the stoichiometric molar ratio can be as high as 34).

Shigeyasu et al. is silent with regard to zirconium. Shigeyasu et al. disclose that too high a temperature, too much cobalt, too high a manganese to cobalt ratio, or too high an absolute amount of manganese results in "a large amount of carbon dioxide gas [being] generated by decomposition of the lower aliphatic carboxylic acid as the solvent, it is not advantageous from an economical standpoint." (Col. 4 lines 1-14, 25-55, Col. 6, lines 7-13). Shigeyasu et al. do not disclose or suggest any particular solvent burn ratio. Shigeyasu et al. do not disclose or suggest that a stoichiometric ratio of bromine to manganese of 1.5 or less reduces the extent of solvent burned relative to other catalyst compositions with the same metal atoms but with molar quantities of bromine in excess of 1.5 with respect to manganese.

The combination of Partenheimer et al. with Shigeyasu et al. does not suggest a catalyst system in the substantial absence of zirconium atoms because Partenheimer requires the presence of zirconium. Following the teachings of the references, the combination of Partenheimer et al. with Shigeyasu et al. would result in a catalyst system containing zirconium.



The combination of Partenheimer et al. with Shigeyasu et al. does not suggest a stoichiometric molar ratio of bromine atoms to manganese atoms of 1.5 or less because at best Partenheimer is silent on the suitable ratio of bromine to manganese, and if calculated from its disclosure, is at least 1.9 (outside the claim element range of 1.5 or less), and Shigeyasu et al. also disclose a bromine to manganese ratio exceeding 1.5.


In sum, neither Partenheimer et al. nor Shigeyasu et al., individually or in combination, disclose or suggest at least two elements of Applicant's claimed invention: a catalyst substantially free of zirconium with a stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less.. Applicant's respectfully note that a prima facie case of obviousness which requires that the prior art references when combined must teach or suggest all claim limitations has not been met since neither Partenheimer et al. nor Shigeyasu et al. nor the combination of the references teach or suggest a process for the oxidation of p-xylene to terephthalic acid comprising a catalyst substantially free of zirconium comprising nickel, manganese, and bromine wherein the stoichiometric molar ratio of bromine atoms to manganese atoms is 1.5 or less.

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71632

In summary, applicant believes the application to be in condition for allowance. Accordingly, the Examiner is respectfully requested to reconsider the rejection(s), enter the above amendment, remove all rejections, and pass the application to issuance.

Respectfully submitted,

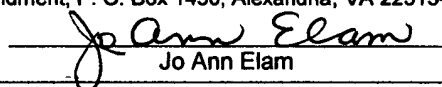
  
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